NORTH ATLANTIC AEROSOL PROPERTIES AND DIRECT RADIATIVE EFFECTS: KEY RESULTS FROM TARFOX AND ACE-2

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1. Introduction

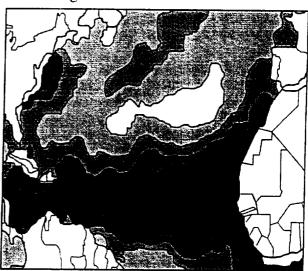
Aerosol effects on atmospheric radiative fluxes provide a forcing function that can change the climate in potentially significant ways. This aerosol radiative forcing is a major source of uncertainty in understanding the observed climate change of the past century and in predicting future climate. To help reduce this uncertainty. the International Global Atmospheric Chemistry Project (IGAC) has endorsed a series of multiplatform aerosol field campaigns. The Tropospheric Radiative Forcing Observational Experiment (TARFOX) and the second Characterization Experiment (ACE-2) were the first IGAC campaigns to address the impact of anthropogenic aerosols.

Both TARFOX and ACE-2 extensive data sets on aerosol properties and radiative effects. TARFOX focused on the urban-industrial haze plume flowing from the eastern United States over the western Atlantic Ocean [Russell et al., 1999a], whereas ACE-2 studied aerosols carried over the eastern Atlantic from both European urban/industrial and African mineral sources [Raes and Bates, 1999]. These aerosols often have a marked influence on the top-of-atmosphere radiances measured by satellites, as illustrated in Figure 1. Shown there are contours of aerosol optical depth derived from radiances measured by the AVHRR sensor on the NOAA-11 satellite. The contours readily show that aercsols originating in North America, Europe, and Africa impact the radiative properties of air over the North Atlantic. However, the accurate derivation of flux changes, or radiative forcing, from the satellite-measured radiances or retrieved optical depths remains a difficult challenge. In this paper we summarize key initial results from TARFOX and, to a lesser extent, ACE-2, with a

focus on those results that allow an improved assessment of the flux changes caused by North Atlantic aerosols at middle and high latitudes.

2. TARFOX Measurements and Calculations

TARFOX made coordinated measurements from four satellites (GOES-8, NOAA-14, ERS-2, LANDSAT), four aircraft (ER-2, C-130, C-131A, and a modified Cessna), land sites, and ships. Included were measurements of aerosolinduced flux changes, made simultaneously with measurements of the chemical, physical, and optical properties of the aerosols causing those changes.



Equivalent Aerosol Optical Depth

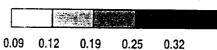


Figure 1. June/July/August map of aerosol optical depth derived from NOAA/AVHRR satellite reflectance data over the oceans [*Husar et al.*, 1997]

Key initial results from TARFOX include:

- Carbonaceous material (organic and inorganic) and sulfates accounted for essentially all of the independently derived total dry aerosol mass in samples obtained at altitudes between 0.2 and 3.8 km [Hegg et al., 1997].
- Averaged over the aerosol layer depth (typically ~3 km), carbonaceous material contributed 50% of dry aerosol mass and 66% of dry aerosol light scattering [Novakov et al., 1997; Hegg et al., 1997]. Both these fractions increased with altitude; fractions near the surface were similar to some other results for the U.S. and Canadian east coasts.
- The mean midvisible single-scattering albedo of the dry aerosol was 0.9, with a range of 0.76 to 0.98 [Hegg et al., 1997].
- The aerosol humidification factor (ratio of light scattering at 80% relative humidity (RH) to that at 30% RH) in the haze plume was on average greater than values previously used in aerosol climate effect calculations, and exceeded the value for sulfate 70% of the time [Kotchenruther et al., 1999].
- Aerosol optical depths derived from in situ scattering and absorption profiles (using measured humidification factors) averaged ~70-90% of airborne sunphotometer values [Hegg et al., 1997; Ross et al., 1998].
- Chemical apportionment of aerosol optical depths showed the dominant importance of water condensed on aerosol, with carbonaceous material second, and sulfate a close third [Hegg et al., 1997].
- Remarkable year-to-year consistency was observed in the characteristics of the aerosol in the TARFOX region during episodes of heavy haze. [Remer et al., 1999].
- Agreement was obtained between aerosol optical thickness magnitudes and wavelength dependences obtained by the MODIS Airborne Simulator (MAS) on the ER-2 and by sunphotometers on the C-131A and the surface [Tanre et al., 1999].
- First validation of a dual-view algorithm that retrieves two-wavelength aerosol optical depth over land and ocean from radiances measured by the Along-Track Scanning Radiometer-2

- (ATSR-2) on the European ERS-2 Satellite [Veefkind et al., 1998, 1999].
- Agreement between shortwave radiative flux changes measured from aircraft and those calculated from measured aerosol properties, provided the calculations use midvisible single scattering albedos for the ambient (humidified) aerosol of ~0.89 to 0.93 [Hignett et al., 1999; Russell et al., 1999b]. An example of this agreement is shown in Figure 2. These single scattering albedo values are consistent with the independent measurements of the dry TARFOX aerosol particles by Hegg et al. [1997] and Novakov et al. [1997], cited above.

3. ACE-2 Measurements and Preliminary Calculations

Early results from ACE-2 have been presented at special sessions of the IGAC conference in Seattle (August 1998) and the Fifth International Aerosol Conference in Edinburgh (September 1998); many are included in several papers at this Symposium [e.g., Schmid et al., 1999]. These results show some similarities between ACE-2 and TARFOX results for the polluted marine boundary layer aerosol, as well as some significant differences. An example of a significant difference is:

 ACE-2 measurements yield consistently smaller organic aerosol mass fractions than were obtained in TARFOX and in some other measurements on the U.S. and Canadian East coasts.

Examples of similarities include:

- Sunphotometer-derived aerosol optical depths usually exceeded values derived from aircraftmeasured profiles of scattering and absorption coefficients, humidification factor, and/or size distribution [e.g., Schmid et al., 1999].
- Aerosol optical depths (AODs) retrieved from AVHRR satellite radiances agreed with values measured by A/C and ship sunphotometers when African dust was absent. However, agreement degraded when African dust was present [e.g., Durkee et al., 1998; Livingston et al., 1998; Schmid et al., 1999].

We have applied to ACE-2 data the methods used to compute the TARFOX flux changes shown in Figure 2. The results are preliminary, in that they use literature values for refractive

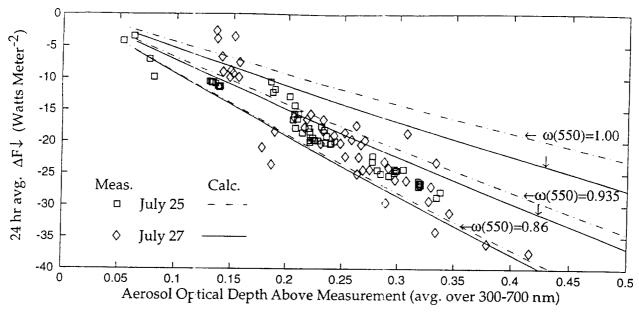


Figure 2. Comparison between aerosol-induced changes in shortwave downward flux determined from TARFOX C-130 measurements (data points) and calculations (curves) for size distributions retrieved from sunphotometer optical depth spectra for two days.

indices of polluted marine boundary-layer and soil-dust aerosols, rather than values based on the ACE-2 size-resolved chemical composition measurements. However, these refractive indices from the literature do yield midvisible single-scattering albedos (~0.94) that are in the range measured for ACE-2 aercsols, both in the polluted marine boundary layer and in elevated African dust layers. The preliminary calculations, which use ACE-2 measured spectra of aerosol optical depth (AOD), yield radiative flux sensitivity (ΔFlux/ΔAOD) values for ACE-2 aerosols (boundary layer and African dust) over the ocean that are similar to values for TARFOX boundary layer aerosols (e.g., $\Delta F \downarrow_{\gamma_{4-hr}}$ $AOD(550 \text{ nm}) \approx -70 \text{ W m}^{-2}$).

4. North Atlantic Flux-Change Calculations

Noting this similarity in flux sensitivities, especially for the polluted boundary-layer aerosols in TARFOX and ACE-2, we have used the aerosol optical model that yielded the agreement in Figure 2 to estimate shortwave radiative flux changes caused by aerosols over the North Atlantic Ocean at middle and high latitudes (i.e., excluding latitudes strongly impacted by African dust aerosols). Our calculations use the North Atlantic AOD values obtained over a two-year period from AVHRR radiances by *Husar et al.* [1997; cf. Figure 1]. Results will be shown for seasonal and annual-average flux changes. To illustrate the

sensitivity of flux changes to aerosol absorption, we show results both for the TARFOX best-fit single-scattering albedo, $\omega(550 \text{ nm})=0.9$, and for non-absorbing aerosols, $\omega=1$. To illustrate the sensitivity to clouds, we show results calculated without clouds and with cloud fractional occurrence from the ISCCP data base.

References

Durkee, P. A., K. E. Nielsen, P. B. Russell, P. B., B. Schmid, J. M. Livingston, D. Collins, R. Flagan, J. Seinfeld, K. Noone, S. Gasso, D. Hegg, T. S. Bates, and P. K. Quinn, Regional aerosol properties from satellite observations: ACE-1, TARFOX and ACE-2 results. J. Aerosol Sci. 29 (Suppl 1): S1149, 1998.

Hegg, D. A., J. Livingston, P. V. Hobbs, T. Novakov, and P. B. Russell, Chemical apportionment of aerosol column optical depth off the Mid-Atlantic coast of the United States, J. Geophys. Res, 102, 25,293-25,303, 1997.

Hignett, P., Taylor, J. P., Francis, P. N., and Glew, M. D., Comparison of observed and modelled direct aerosol forcing during TARFOX, J. Geophys. Res in press, 1999.

Husar, R. B., J. M. Prospero and L. L. Stowe, Characterization of tropospheric aerosols over the oceans with the NOAA advanced very high resolution radiometer optical thickness operational product, *J. Geophys. Res.*, 102, 16,889-16,909, 1997.

Kotchenruther, R.A., P. V. Hobbs, and D. A. Hegg, Humidification factors for aerosols off the mid-Atlantic coast of the United States, J.

Geophys. Res., in press, 1999.

Livingston, J. M., V. Kapustin, B. Schmid, P. B. Russell, P. A. Durkee, T. S. Bates, and P. K. Quinn, "Shipboard sunphotometer measurements of aerosol optical depth spectra and columnar water vapor during ACE-2, J. Aerosol Sci. 29 (Suppl 1): S257-S258, 1998.

Novakov, T., D. A. Hegg and P. V. Hobbs, Airborne measurements of carbonaceous aerosols on the east coast of the United States, J. Geophys. Res., 102, 30,023-30,030,

1997.

Raes, F., T. Bates, G. Verver, D. Vogelenzang, M. Van Liedekerke, The second Aerosol Characterization Experiment (ACE-2): Introduction, meteorological overview and main results, *Tellus*, submitted, 1999.

Remer, L. A., Y. J. Kaufman and B. N. Holben, Interannual variation of ambient aerosol characteristics on the east coast of the United States, *J. Geophys. Res.*, in press, 1999.

- Ross, J., P. V. Hobbs, W. S. Hartley, J. M. Livingston, and P. B. Russell, Use of airborne measurements and aerosol closure tests to derive a model for the optical properties of aerosols off the East coast of the United States, J. Geophys. Res., submitted, 1998.
- Russell, P. B., P. V. Hobbs, and L. L. Stowe, Aerosol properties and radiative effects in the United States Mid-Atlantic haze plume: An overview of the Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX), J. Geophys. Res., in press, 1999a.

Russell, P. B., J. M. Livingston, P. Hignett, S. Kinne, J. Wong, A. Chien, R. Bergstrom, P. Durkee, and P. V. Hobbs, Aerosol-induced radiative flux changes off the United States Mid-Atlantic coast: Comparison of values calculated from sunphotometer and in situ data with those measured by airborne pyranometer, J. Geophys. Res., in press, 1999b.

Schmid, B., P. B. Russell, J. M. Livingston, S. Gasso, D. A. Hegg, D. R. Collins, R. C. Flagan, J. H. Seinfeld, E. Ostrom, K. J. Noone, P. A. Durkee, H. Jonsson, E. J. Welton, K. Voss, H. R. Gordon, P. Formenti, M. O. Andreae, V. N. Kapustin, T. S. Bates, and P. K. Quinn, Clear column closure studies of urban-marine and mineral-dust aerosols using aircraft, ship, satellite and ground-based measurements in ACE-2, ALPS 99 Symposium, Meribel, France, 18-22 January 1999.

Tanré, D., L.A. Remer, Y.J. Kaufman, S. Mattoo, P.V. Hobbs, J.M. Livingston, P.B. Russell, A. Smirnov, Retrieval of aerosol optical thickness and size distribution over ocean from the MODIS Airborne Simulator during TARFOX, J. Geophys. Res., in press,

1999.

Veefkind, J. P., de Leeuw, G., Durkee, P. A., Retrieval of aerosol optical depth over land using two-angle view satellite radiometry during TARFOX, *Geophys. Res. Lett.* 25, 3135-3138, 1998.

Veefkind, J.P., G. de Leeuw, P.A. Durkee, P.B. Russell, and P. V. Hobbs, Aerosol optical depth retrieval using ATSR-2 and AVHRR data during TARFOX, J. Geophys. Res., in press, 1999.